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Indirect N_2O emissions with seasonal variations from an agricultural drainage ditch mainly receiving interflow water☆

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ABSTRACT

Nitrogen (N)-enriched leaching water may act as a source of indirect N_2O emission when it is discharged to agricultural drainage ditches. In this study, indirect N_2O emissions from an agricultural drainage ditch mainly receiving interflow water were measured using the static chamber-gas chromatography technique during 2012–2015 in the central Sichuan Basin in southwestern China. We found the drainage ditch was a source of indirect N_2O emissions contributing an inter-annual mean flux of 6.56 ± 1.12 μg N m^{-2} h^{-1} and a mean indirect N_2O emission factor (EF_{5g}) value of 0.03 ± 0.003%. The mean EF_{5g} value from literature review was 0.51%, which was higher than the default EF_{5g} value (0.25%) proposed by the Intergovernmental Panel on Climate Change (IPCC) in 2006. Our study demonstrated that, more *in situ* observations of N_2O emissions as regards N leaching are required, to account for the large variation in EF_{5g} values and to improve the accuracy and confidence of the default EF_{5g} value. Indirect N_2O emissions varied with season, higher emissions occurred in summer and autumn. These seasonal variations were related to drainage water NO_3^--N concentration, temperature, and precipitation. Our results showed that intensive precipitation increased NO_3^--N concentrations and N_2O emissions, and when combined with warmer water temperatures, these may have increased the denitrification rate that led to the higher summer and autumn N_2O emissions in the studied agricultural drainage ditch.

1. Introduction

Atmospheric concentrations of the ozone-depleting, and potent greenhouse gas nitrous oxide (N_2O), have increased from a pre-industrial level of 270 ppb–324 ppb in 2011. One of the main causes of this rise in atmospheric N_2O is the increasing use of nitrogen (N) fertilizers (Ravishankara et al., 2009; IPCC, 2013). Considerable amounts of N are lost from N-fertilized agricultural land via leaching and runoff, and which is ultimately transported into groundwater, drainage ditches, rivers and estuaries, consequently causing N pollution in aquatic ecosystems (Mosier et al., 1998; Mulholland et al., 2008; Zhu et al., 2009; Gumiero et al., 2011). There have been several reports of groundwater contaminated with high nitrate (NO_3^-) concentrations in areas of high fertilizer use (Groffman et al., 1998; McMahon et al., 2000; Hiscock et al., 2003; Jahangir et al., 2013). In addition to studies on direct emissions of N_2O from N fertilized soils, indirect N_2O emissions from aquatic ecosystems that are associated with N leaching and runoff in agricultural areas deserve attention (Nevison, 2000; Beaulieu et al., 2008; Outram and Hiscock, 2012; Tian et al., 2017). The N-enriched groundwater associated with N leaching is considered a source of indirect N_2O emissions via denitrification or degassing when it is discharged to adjacent watercourses such as drainage ditches and streams (McMahon et al., 2000; Reay et al., 2004a, 2004b; Minamikawa et al., 2011; Jurado et al., 2017). Werner et al. (2012), for example, found that agricultural streams were a significant source of N_2O, while Jurado et al. (2017) also found that groundwater could act as a source of N_2O to the atmosphere and with the highest level of N_2O flux in springs supplied by groundwater compared with those in wetland and estuarine areas. Reay et al. (2004a and 2004b) reported that concentrations of dissolved N_2O

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in leachate rapidly decreased on entry to drainage ditches.

Drainage ditches in farmlands are generally ubiquitous and, as such, represent important hydrologic conduits for surface and subsurface N flow to aquatic systems (Kröeger et al., 2007; Shen et al., 2016; Zhu et al., 2012). Many drainage ditches are polluted and suffer from eutrophication, owing to losses of N from agriculture (Janse and Van Puijenbroek, 1998). But drainage ditches also act as important sites for biogeochemical interactions between reactive N, aquatic plants, microorganisms, and the physical environment (Janse and Van Puijenbroek, 1998; Shen et al., 2016; Zhang et al., 2016). Consequently, drainage ditches have been identified as hotspots for N removal and N_2O emissions (Reay et al., 2003, 2004a; Kröeger et al., 2007; Zhang et al., 2016). It is possible that spatial and temporal variations in N_2O emissions can be caused by dynamic changes in drainage water NO_3^- concentrations and other geochemical and hydrological parameters (Beaulieu et al., 2009; Jurado et al., 2017; Tian et al., 2017). It has thus been suggested that these variations should be considered in improving the certainty of quantification of indirect N_2O emissions (Werner et al., 2012; Jurado et al., 2017).

The Intergovernmental Panel on Climate Change (IPCC) has defined the emission factor for indirect N_2O emissions from leaching and runoff from agricultural systems as EF_5. This EF_5 incorporates three components: EF_{5g}, EF_{5r} and EF_{5e}, which are the emission factors for groundwater and surface drainage, rivers, and estuaries, respectively (IPCC, 2006). The default value of the EF_5 was defined as the proportion of N leaching and runoff converted to N_2O in these water bodies (IPCC, 2006). However, the default value proposed by the IPCC to estimate N_2O emissions in drainage ditches and groundwater resulting from leached N has a lack of certainty (Clough et al., 2007a; Beaulieu et al., 2008; Outram and Hiscock, 2012; Jahangir et al., 2013), since it has decreased from 2.5% in 1997 to 0.25% in 2006, based on studies from a limited number of countries (IPCC, 2006; Outram and Hiscock, 2012). In view of the large variation (0.002%–73%) in the values of EF_{5g} (Jurado et al., 2017), the default value requires improvement by increasing the number of global *in situ* observations (Reay et al., 2003; Beaulieu et al., 2008; Outram and Hiscock, 2012).

In China, the sloping farmland of the purple soils in the central Sichuan Basin is particularly vulnerable to N loss via NO_3^--N leaching due to a combination of intensive farming practices, hilly topography, climate, and soil characteristics (Zhu et al., 2009; Wang and Zhu, 2011; Gao et al., 2014), where the annual loss of N from these soils via interflow was reported to be 37.9 kg ha^{-1}, and accounted for 88% of the total N loss (Zhu et al., 2009). Interflow is the lateral movement of water in the unsaturated zone, that returns to the surface or enters a stream prior to becoming groundwater. Interflow was reported as an important water flow pattern in this area (Zhao et al., 2013), where the main N loss pathway from the local farmland is NO_3^--N leaching via interflow (Wang and Zhu, 2011). Previous studies have also reported that interflow was the predominant pathway of water discharge (Zhu et al., 2009; Zhao et al., 2013; Hua et al., 2014) and a primary source of water for shallow groundwater recharge, ditches, and streams in this area (Wang and Zhu, 2011; Zhao et al., 2013). This region is a nitrate sensitive area, because the loading of nitrate leaching in the purple soil area is more than 2-fold the average loss in China (Zhou et al., 2013; Zhang et al., 2013). There is severe NO_3^--N pollution of groundwater (mean concentration of NO_3^--N >10 mg L^{-1}) and water eutrophication in the region (Zhu et al., 2009; Wang and Zhu, 2011; Zhou et al., 2013). Moreover, the long-distance movement of N discharged from the purple soil area may have a profound impact on the water quality of the nearby Yangtze River (Wang and Zhu, 2011).

Indirect N_2O emissions from an agricultural drainage ditch mainly receiving interflow were measured for three years *in situ* in southwestern China in this study. The objectives of this study were to quantify the indirect N_2O emissions from agricultural drainage ditches, examine the temporal variation in N_2O emissions, and explore the factors affecting the indirect N_2O emissions, since little is known about the indirect N_2O emissions and EF_{5g} from drainage ditches mainly receiving interflow water in this area.

2. Materials and methods

2.1. Study area

The field study was carried out at the Yanting Agro-Ecological Station of Purple Soil (N 31°16′, E 105°28′), a station of the Chinese Ecosystem Research Network (CERN), Chinese Academy of Sciences (CAS), in an important agricultural area in the upper tributary of the Yangtze River Watershed (Fig. 1a and b). Altitude at the study area ranges between 400 and 600 m, and the surface is mainly covered by low mountains, and hills. The area has a humid subtropical monsoon climate, with an annual (1981–2009) mean temperature of 17.3°C and seasonally variable precipitation of 836 mm spring: 5.9%; summer: 65.5%; autumn: 19.7%; and, winter: 8.9%; from 1981 to 2006 data, after Zhu et al. (2009).

The drainage ditches were located in the valley bottom of a small agricultural catchment (0.15 km^2; Fig. 1b) of the first-order tributary of the Yangtze River (Zhu et al., 2012), where the land use was dominated by sloping farmland of purple soil and forest. The soil is classified as a Regosol (FAO Soil Taxonomy) or a Pup-Orthic-Entisol (Chinese Soil Taxonomy) (Zhu et al., 2012). Land use distribution reflected the topography, with paddy fields on low-lying parts of hills, and farmland on slopes ranging from 3° to 15°. Forestry is mainly concentrated on upper parts of the hills. Rice (*Oryza sativa* L.) is cultivated in the paddies in the rainy season (from the middle of May to September) with applications of 150 kg N ha^{-1}, while oilseed rape (*Brassica napus* L.) is cultivated in the dry season (from late October to early May) with an application of 130 kg N ha^{-1} in late October. Maize (*Zea mays* L.) is planted on sloping farmland in the rainy season and winter wheat (*T. aestivum* L.) is cultivated in the dry season, with applications of 150 kg N ha^{-1} and 130 kg N ha^{-1}, respectively. Forestry is dominated by Alder, *Alnus cremastogyne* Burk., and Cypress (*Cupressus funebris* Endl) plantations. The drainage ditch was surrounded by the upland farmlands, vegetable fields and paddy rice fields (Fig. 1b). The width and depth of the drainage ditch was ~70 cm and 70–100 cm, respectively. The water depth in the ditch, measured using a stainless steel ruler, was shallow (0.6–6.4 cm) with a slow velocity during the observation period, and with sediment depth < 20 cm. The ditch was artificially excavated, and the purplish shale and soil layer interface was exposed to help leaching water laterally flow into the drainage ditch, since the bedrock has extremely weak permeability (Zhu et al., 2009; Zhao et al., 2013, Fig. 1c). Moreover, the local groundwater level is usually very shallow (−45.5–−193.2 cm, Zhang et al., 2017), and the ditch receives shallow groundwater recharge and leaching water for most of the year, while it also receives overland flow for a short period of time (1–2 days) after intensive precipitation. Thus, this drainage ditch receives interflow as the main water source. During the early spring (dry season), there is no water in the drainage ditch for several days. Vegetation in the study ditch mainly comprised *Lolium perenne* L., *Echinochloa crus-galli* (L.) Beauv., *Fimbristylis milliacea* (L.) Vahl, *Polygonum hydropiper*, and other ruderal weeds.

2.2. Sample collection and analysis

The N_2O emissions from drainage ditch were measured *in situ* by

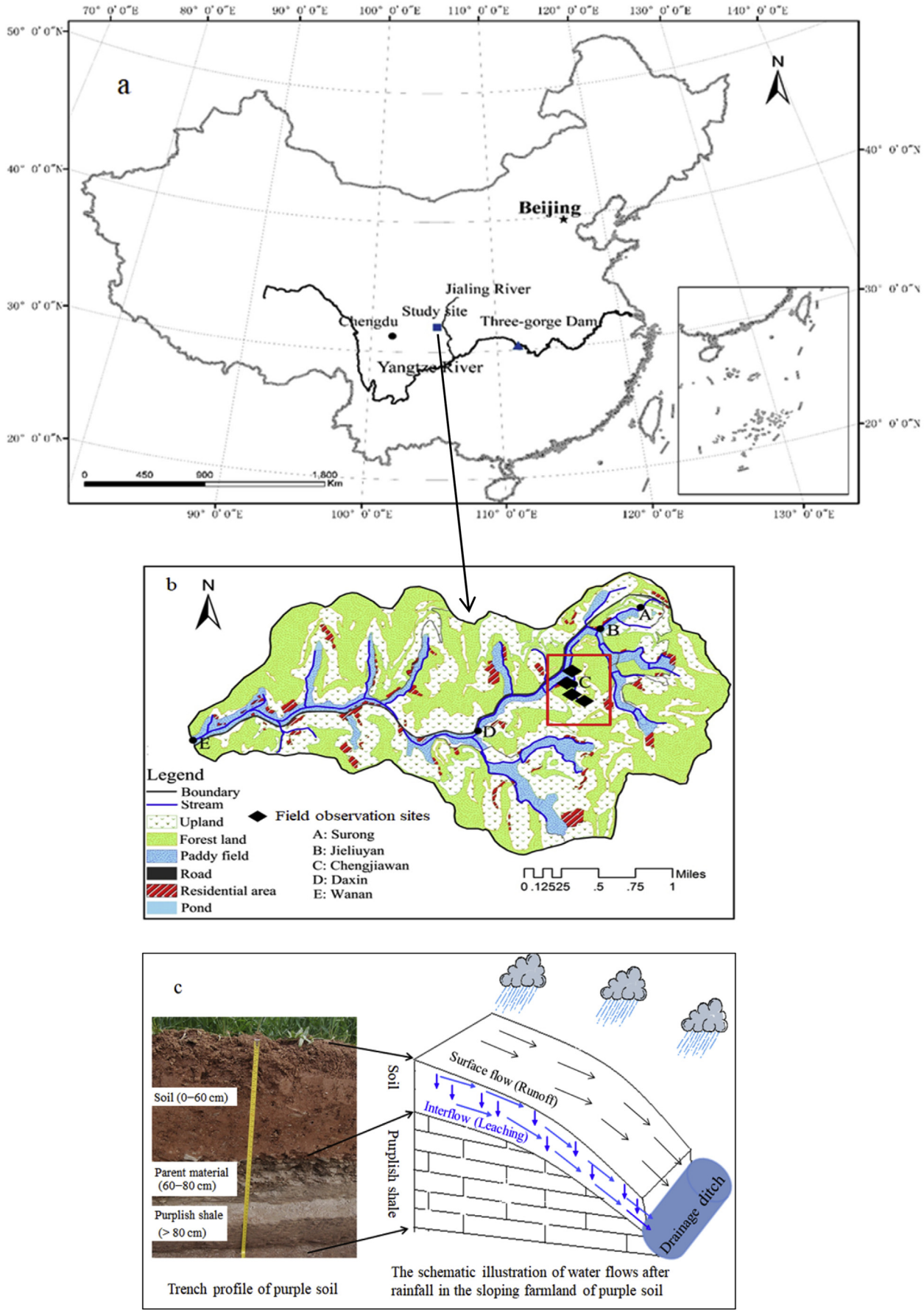

Fig. 1. **(a)** Location and **(b)** catchment of the study site in southwestern China; **(c)** profile of purple soil (left) and schematic illustration of post-rainfall water flow movement in the study site purple soil type (right); and **(d)** schematic diagram of the gas collection device in the field. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

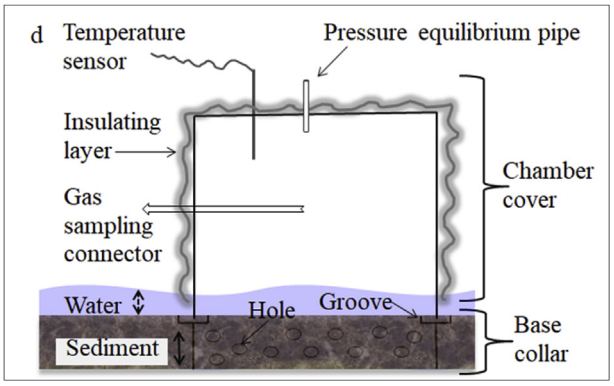

Fig. 1. (*continued*).

employing the static chamber-gas chromatography technique. However, this technique may cause artefacts in determining N_2O emission due to the effect of water turbulence around the chamber (Clough et al., 2011). Nonetheless, the floating chamber method could be relatively simple and rapid methodology for observation of representative N_2O emissions from rivers (Xia et al., 2014), and the N_2O fluxes measured by floating chamber might be slightly higher than those calculated from N_2O concentration data (Harrison and Matson, 2003). However, the water depth of the drainage ditch in the present study was quite shallow and unsuitable for using the floating chamber method to measure N_2O emission flux. Thus the static chamber technique was used in this study, where four chambers were installed at 25–30 m intervals around the interflow discharge outlet spanning a length of 100 m. The chambers contained two parts: a chamber cover measuring 50 cm × 50 cm × 50 cm as a five-plane cube, and a square base collar measuring 50 cm long × 50 cm wide × 10 cm high with a 3 cm × 3 cm groove on the top. The materials and structures of the two parts of chambers were previously reported (Tian et al., 2017). The base collars were inserted into the ditch sediment at a depth of ~10 cm (Fig. 1d), and kept in place for the one-year observation period, before they were moved to near the original sampling point in the following year. There were some holes (Φ 2.0 cm) equally distributed in the side walls of the base collar to allow the lateral transfer of water plants, animals and microbes, and sediment nutrients (Tian et al., 2017). When gases were sampled, the chamber covers were temporarily placed onto the base collar and the holes in the side walls of the base collars were submerged in drainage water to avoid gas leakage. Gas samples were collected between 9:00–11:00 a.m., 1–2 times per week from 1st December to 30th November in 2012–2013, 2013–2014, and 2014–2015. On each sampling occasion, five × 50 mL of chamber air were collected at 0, 7, 14, 21, and 28 min after closing the chamber, using a plastic syringe mounted with a three-way valve. The sample N_2O concentrations were measured within 24 h after gas collection, using a gas chromatograph (Agilent 7890, Santa Clara, CA, USA) fitted with an electron capture detector (ECD) that was operated at 330 °C. The depth of water and water temperature (WT) at 0–5 cm depth, air temperature in the chamber and pH (in 2014–2015 only) were recorded concurrently with gas emission sampling. The water dissolved oxygen (DO) concentrations in the ditch were only measured *in situ* during the second half of the third observation year, because of the late arrival of purchased instrument for measuring DO. In 2013–2014 and 2014–2015, 500 mL of water was collected from each gas emissions recording site and transported in an insulated box filled with ice-packs to the laboratory, where the samples were stored at 4°C until analysis for inorganic N and dissolved organic carbon (DOC) concentrations using methods reported by Tian et al. (2017). Meteorological data (daily precipitation, air temperature and barometric pressure) were obtained from the meteorological station at the Yanting Agro-Ecological Station of Purple Soil of CERN, located approximately 1 km away from the study site.

2.3. Data analysis

The N_2O fluxes (μg N m^{-2} h^{-1}) were calculated from the increases in N_2O concentrations of the five gas samples within the measurement period (Tian et al., 2017). The cumulative N_2O emissions (kg N ha^{-1}) were derived from the flux calculations using a linear interpolation method (Zhou et al., 2013, 2015; Tian et al., 2017). Although linear interpolation method is commonly used in previous studies, it has some uncertainties in estimating N_2O emission, because this method might miss short-lived N_2O emission peaks, given the episodic nature of indirect N_2O fluxes. We calculated the EF_{5g} value from the mass ratio of emitted N_2O-N to NO_3^--N in a unit volume of drainage water (Well et al., 2005b; IPCC, 2006). We averaged all EF_{5g} in each observation season and annual year for the mean values of seasonal and annual EF_{5g}.

In our study area, winter starts from December to the end of February, summer includes June, July and August, spring and autumn are during the transitions between winter and summer. The inter-annual and seasonal variations in N_2O emissions and mean values of drainage water variables were assessed using one-way ANOVA, followed by Hochberg's GT2 multiple comparison test ($P < 0.05$). Using simple linear regression, we tested for relationships between drainage water NO_3^--N concentration and the accumulated amount of intensive rainfall (>15 mm) that fell 3 days prior to measuring water variables, cumulative N_2O emissions and accumulated rainfall during each rainfall event, and drainage water NO_3^--N concentration following each rainfall event in 2014–2015. Only the accumulated amount of precipitation >15 mm were used for the analysis, i.e., when the drainage water NO_3^--N concentration was measured on 24th May, the accumulated precipitation 3 days prior to measuring NO_3^--N was cumulative rainfall from 21st to 22nd May. Emission of N_2O during each rainfall event defined as cumulative N_2O emission during the rainy days and subsequent 2 days for probable N leaching, i.e., when 21st to 22nd May were rainy days, the period for cumulative N_2O emission was from 21st May to 24th May. The relationships between ln (mean seasonal N_2O flux) and seasonal precipitation, mean seasonal air and water temperature were also analyzed using simple linear regression analysis, which was also used for those relationships analysis on monthly scale. Data for monthly and seasonal N_2O fluxes were ln transformed and statistical analyses were performed using SPSS version 20.0 (SPSS, Inc., USA).

Table 1
Seasonal variation in drainage water variables, indirect N_2O fluxes, and indirect N_2O emission factor (EF_{5g}) and mean annual indirect N_2O fluxes.

Observation period	Drainage water variable	Mean annual value	Mean seasonal value			
			Winter	Spring	Summer	Autumn
Dec 2012–Nov 2013	Water temperature (°C)	17.0	6.96 ± 0.62 a	21.0 ± 0.70 b	24.2 ± 0.39 b	21.4 ± 0.36 b
	N_2O flux (μg N m^{-2} h^{-1})	4.72 ± 2.85	2.13 ± 0.11 A	4.06 ± 0.71	6.80 ± 4.65	5.24 ± 3.98
Dec 2013–Nov 2014	NH_4^+–N (mg N L^{-1})	0.13	0.09 ± 0.02	0.07 ± 0.02	0.18 ± 0.02	0.15 ± 0.04
	NO_3^--N (mg N L^{-1})	3.17	3.53 ± 0.27 b	1.74 ± 0.15 ab	1.49 ± 0.23 a	9.32 ± 1.24 c
	DOC (mg C L^{-1})	2.17	1.31 ± 0.10 a	1.67 ± 0.32 a	3.09 ± 0.29 b	2.30 ± 0.48 ab
	Water temperature (°C)	15.3	6.50 ± 0.44 a	9.29 ± 0.58 a	23.3 ± 0.37 b	21.7 ± 0.63 b
	EF_{5g}	0.017%	0.004% a	0.006% a	0.035% b	0.004% a
	N_2O flux (μg N m^{-2} h^{-1})	5.20 ± 2.23	0.97 ± 1.10 Aa	2.51 ± 2.16 a	7.06 ± 2.52 b	7.71 ± 2.92 b
Dec 2014–Nov 2015	NH_4^+–N (mg N L^{-1})	0.09	0.10 ± 0.01	0.13 ± 0.06	0.10 ± 0.01	0.08 ± 0.01
	NO_3^--N (mg N L^{-1})	4.09	1.56 ± 0.29 a	1.16 ± 0.40 a	4.44 ± 1.03 ab	6.54 ± 0.17b
	DOC (mg C L^{-1})	2.30	1.50 ± 0.13 a	4.38 ± 0.68 c	2.88 ± 0.27 b	1.72 ± 0.12 ab
	Water temperature (°C)	17.6	7.74 ± 0.35 a	14.7 ± 0.81 b	23.5 ± 0.22 d	18.9 ± 0.74 c
	pH	7.56	7.68 ± 0.03	7.63 ± 0.27	7.51 ± 0.04	7.48 ± 0.07
	EF_{5g}	0.034%	0.038%	0.045%	0.039%	0.020%
	N_2O flux (μg N m^{-2} h^{-1})	9.77 ± 3.99	5.78 ± 2.30 Ba	4.25 ± 1.27 a	10.02 ± 3.34 b	14.79 ± 7.00 b
Average of three years	N_2O flux (μg N m^{-2} h^{-1})	6.56 ± 1.61	2.96 ± 1.45 a	3.61 ± 0.55 a	7.96 ± 1.03 b	9.25 ± 2.86 b

Values are means ± standard errors (n = 15, 3, 15, and 2 for Winter, Spring, Summer, and Autumn, respectively, in Dec 2012–Nov 2013; n = 13, 4, 14, and 4 for Winter, Spring, Summer, and Autumn, respectively, in Dec 2013–Nov 2014; and, n = 15, 4, 23, and 17 for Winter, Spring, Summer, and Autumn, respectively, in Dec 2014–Nov 2015). Different lowercase letters within a row indicate differences among the seasons ($P < 0.05$), while different capital letters indicate differences among years ($P < 0.05$). Concentrations of NH_4^+, NO_3^- and DOC were measured in Dec 2013–Nov 2014 and Dec 2014–Nov 2015. Observation period for N_2O emissions in Dec 2012–No v, 2013, Dec 2013–Nov 2014, and Dec 2014–Nov 2015 were 284, 287,and 298 days, respectively.

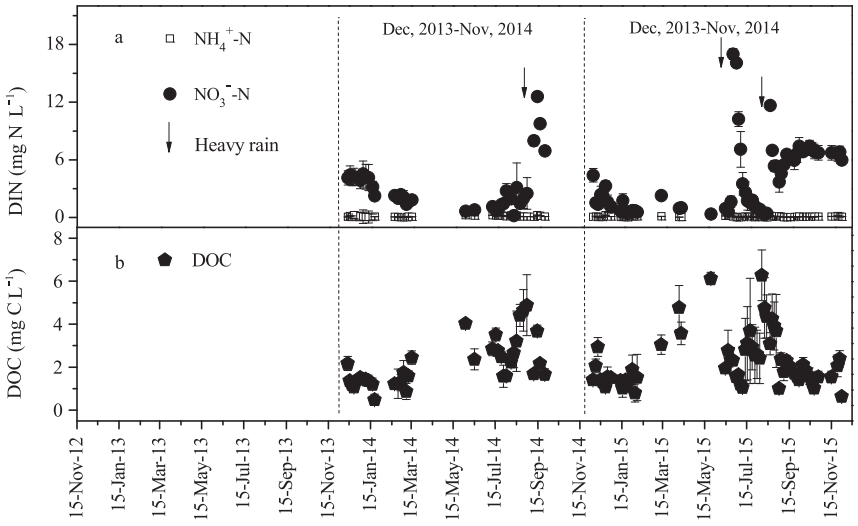

Fig. 2. Concentrations of NH_4^+, NO_3^- and DOC in drainage water during two years of the study. Data points are means and error bars are the standard errors of replicates (n = 4).

3. Results

3.1. Temperature and precipitation

Mean daily air temperature ranged from 0.4 to 30.6 °C during the experimental period, 2012–2015, where mean annual air temperature was 17.3, 16.6, and 17.0 °C in 2012–2013, 2013–2014 and 2014–2015, respectively (Fig. S1a). Mean seasonal air temperature was 6.63, 17.9, 25.8 and 17.3 °C, in winter, spring, summer, and autumn, respectively. Water temperature ranged from 3.4 to 27.4 °C during the three year study period, where mean annual temperature was 17.0, 15.3, and 17.6 °C in 2012–2013, 2013–2014, and 2014–2015, respectively (Fig. S1b). There were seasonal variations in mean water temperature, where it was 7.1, 15.0, 23.7, and 20.7 °C in winter, spring, summer, and autumn, respectively. There was a positive correlation between water temperature and air temperature (r = 0.97, n = 131).

Annual precipitation was 1272, 821, and 956 mm in 2012–2013, 2013–2014, and 2014–2015, respectively (Fig. S1c). The mean seasonal precipitation in winter, spring, summer, and autumn was 18.2, 147, 519, and 333 mm, respectively, where 83.8% of the precipitation occurred in summer and autumn.

3.2. Drainage water

Depth of the drainage ditch water varied from 0.6 cm to 6.4 cm (mean: 1.8 cm), with higher mean seasonal water depths in summer and autumn than those in winter and spring (data not shown). We recorded pH of the water during 2014–2015 and found the mean was 7.56, with no variation among the seasons, and similarly, there was no seasonal variation in NH_4^+–N concentrations (Table 1). Concentrations of NH_4^+–N (range: 0.03–0.31 mg N L^{-1}, mean: 0.11 mg N L^{-1}) were lower ($P < 0.001$) than those of NO_3^--N (range: 0.20–17.0 mg N L^{-1}, mean: 3.75 mg N L^{-1}; Fig. 2a). The mean NO_3^--N concentration in autumn was higher ($P < 0.05$) than those in the other seasons in 2013–2014 and 2014–2015 (Table 1). However, neither concentration of NH_4^+–N nor NO_3^--N varied between 2013–2014 and 2014–2015 ($P > 0.05$; Table 1). Concentrations of

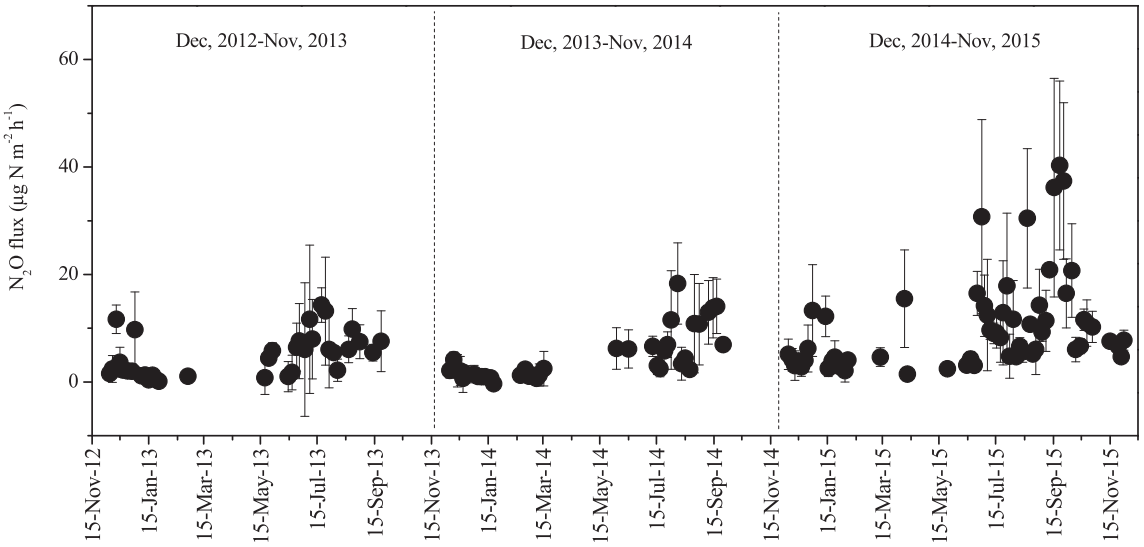

Fig. 3. N_2O fluxes recorded throughout the study period. Data points are means and error bars are the standard errors of replicates ($n = 4$).

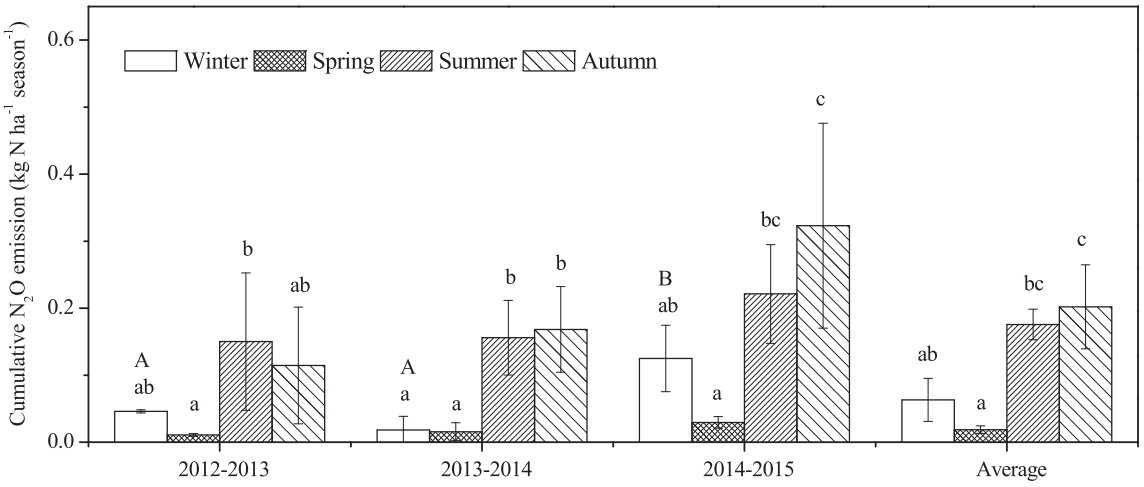

Fig. 4. N_2O emissions in each season over the three years study period. Data points are means and error bars are the standard errors of replicates ($n = 4$). Different capital letters indicate differences among the years, while different lowercase letters indicate differences among the seasons ($P < 0.05$).

DOC ranged from 0.49 to 6.28 mg C L^{-1}, and had a mean value of 2.25 mg C L^{-1} (Table 1; Fig. 2b).

3.3. Indirect N_2O emissions and N_2O emission factor (EF_{5g})

Over the three years, N_2O fluxes ranged from −0.33–40.3 μg N m^{-2} h^{-1} (Fig. 3), with a mean of 6.56 μg N m^{-2} h^{-1} and a cumulative emission of 1.38 kg N ha^{-1} (Table 1). Mean annual N_2O fluxes were 4.72, 5.20, and 9.77 μg N m^{-2} h^{-1} in 2012–2013, 2013–2014, and 2014–2015, respectively (Table 1), and there were no variations in mean annual fluxes or total annual N_2O emissions (Table 1). But the cumulative seasonal N_2O emissions in winter in 2014–2015 were higher than those in winter in 2012–2013 and 2013–2014 (Fig. 4). We also found within-year seasonal differences in each of the three years, where cumulative seasonal N_2O emissions were higher in summer than in spring in 2012–2013 ($P < 0.05$), higher in summer and autumn than in winter and spring in 2013–2014 ($P < 0.05$), and higher in summer and autumn than in spring in 2014–2015 ($P < 0.05$; Fig. 4).

We found that the EF_{5g} values ranged from 0.002% to 0.19%, with a mean of 0.03%, with no variation in value between 2013–2014 and 2014–2015 (Table 1). However, mean EF_{5g} in summer was higher ($P < 0.05$) than those in the other seasons in 2013–2014 (Table 1).

3.4. Relationships between N_2O dynamics and environmental factors

There were positive relationships between the cumulative precipitation (>15 mm) 3 days prior to measuring NO_3^--N and drainage water NO_3^--N concentration measured after precipitation in 2014–2015 ($R^2 = 0.55$, $P < 0.05$, $n = 12$; Fig. 5a), the cumulative precipitation and the cumulative N_2O emissions during rainfall events ($R^2 = 0.35$, $P < 0.05$, $n = 13$; Fig. 5b), and cumulative N_2O emissions during rainfall events and drainage water NO_3^--N concentration measured after rainfall events ($R^2 = 0.55$, $P < 0.05$, $n = 12$; Fig. 5c). There were positive relationships between ln (mean monthly N_2O flux) and mean monthly air temperature ($R^2 = 0.36$, $P < 0.001$, $n = 33$; Fig. 6a), mean monthly water temperature ($R^2 = 0.47$, $P < 0.001$, $n = 33$; Fig. 6b), and monthly cumulative

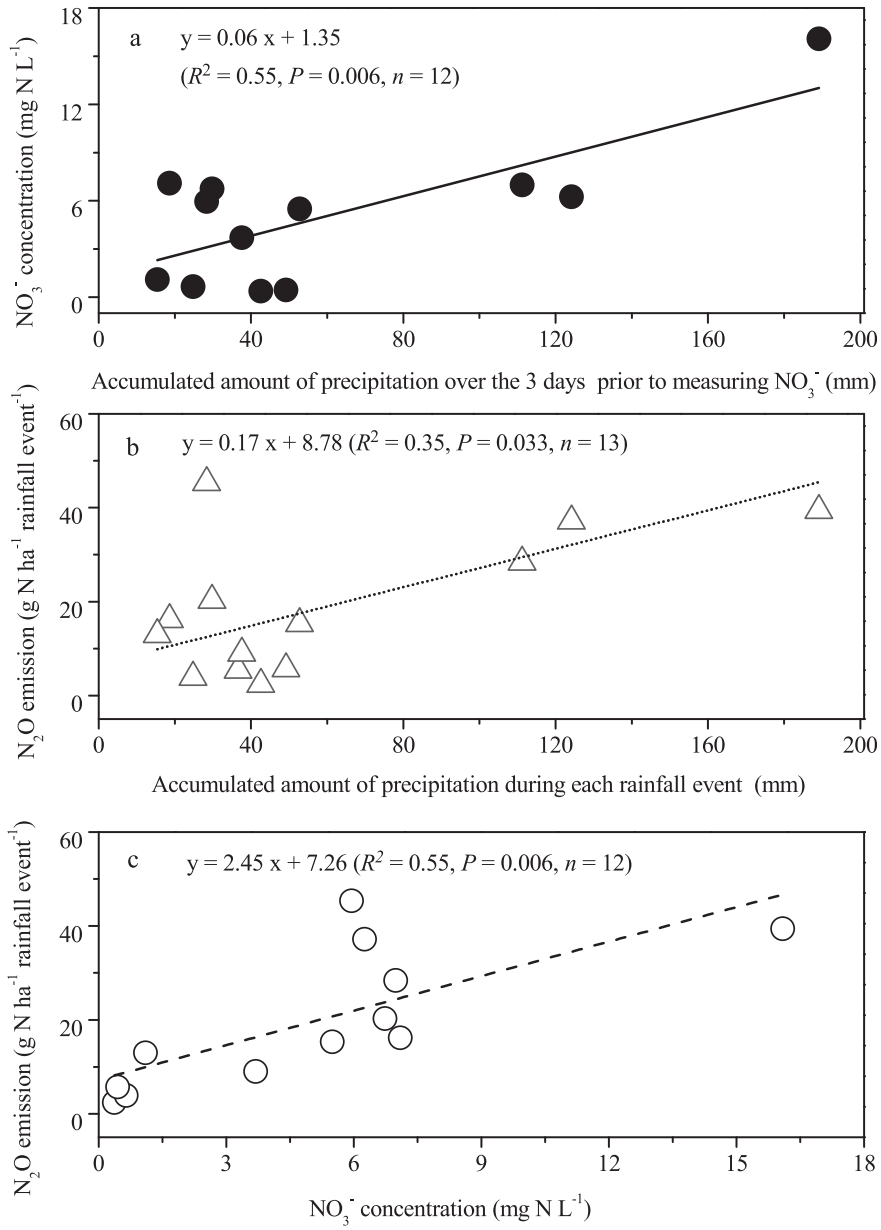

Fig. 5. Regression analyses of **(a)** accumulated amount of precipitation (>15 mm) that fell over the 3 days prior to measuring NO_3^--N and drainage water NO_3^--N concentration following precipitation events; **(b)** N_2O emissions and accumulated amount of precipitation during rainfall events; **(c)** N_2O emissions during each rainfall event and drainage water NO_3^--N concentration after rainfall events.

precipitation ($R^2 = 0.24$, $P < 0.05$, $n = 33$; Fig. 6c). We also found positive relationships between ln (mean seasonal N_2O flux) and mean seasonal air temperature ($R^2 = 0.46$, $P < 0.05$, $n = 12$; Fig. 6d), mean seasonal water temperature ($R^2 = 0.35$, $P < 0.05$, $n = 12$; Fig. 6e), and seasonal cumulative precipitation ($R^2 = 0.53$, $P < 0.05$, $n = 12$; Fig. 6f).

4. Discussion

4.1. Comparison with other studies

When we compared this study with previous studies on indirect N_2O fluxes from groundwater and drainage water (Table 2), we found our result (6.56 μg N m^{-2} h^{-1}) was higher than those from the UK (Aberdeenshire, Reay et al., 2009; Cambridgeshire, Mühlherr & Hiscock, 1997, and Hiscock et al., 2003; Norfolk, Hiscock et al., 2003, and Hama-Aziz et al., 2017), the USA (the High Plains aquifer, McMahon et al., 2000), and France (the Seine Basin, Vilain et al., 2012), but similar to fluxes from drainage water in Japan and Canada (Table 2). But, the value in this study was much lower than those ranging between 35.2 and 7440 μg N m^{-2} h^{-1} recorded from the UK (Midlothian, Reay et al., 2003 and Norfolk, Outram and Hiscock, 2012), the USA (the Kalamazoo River Basin, Beaulieu et al., 2008; the Choptank River and Nanticoke River Basins, Gardner et al., 2016), Italy, and Sweden (Table 2). We also found that, the mean N_2O flux was similar to direct N_2O fluxes (7.53 μg N m^{-2} h^{-1}) from a rice-rapeseed rotation without N fertilization located in the same study area (Zhou et al., 2015). We deduce, therefore, the agricultural drainage ditches mainly receive interflow water represent non-negligible sources of indirect N_2O emission in the central Sichuan Basin in China.

We summarized the EF_{5g} values from peer-reviewed papers

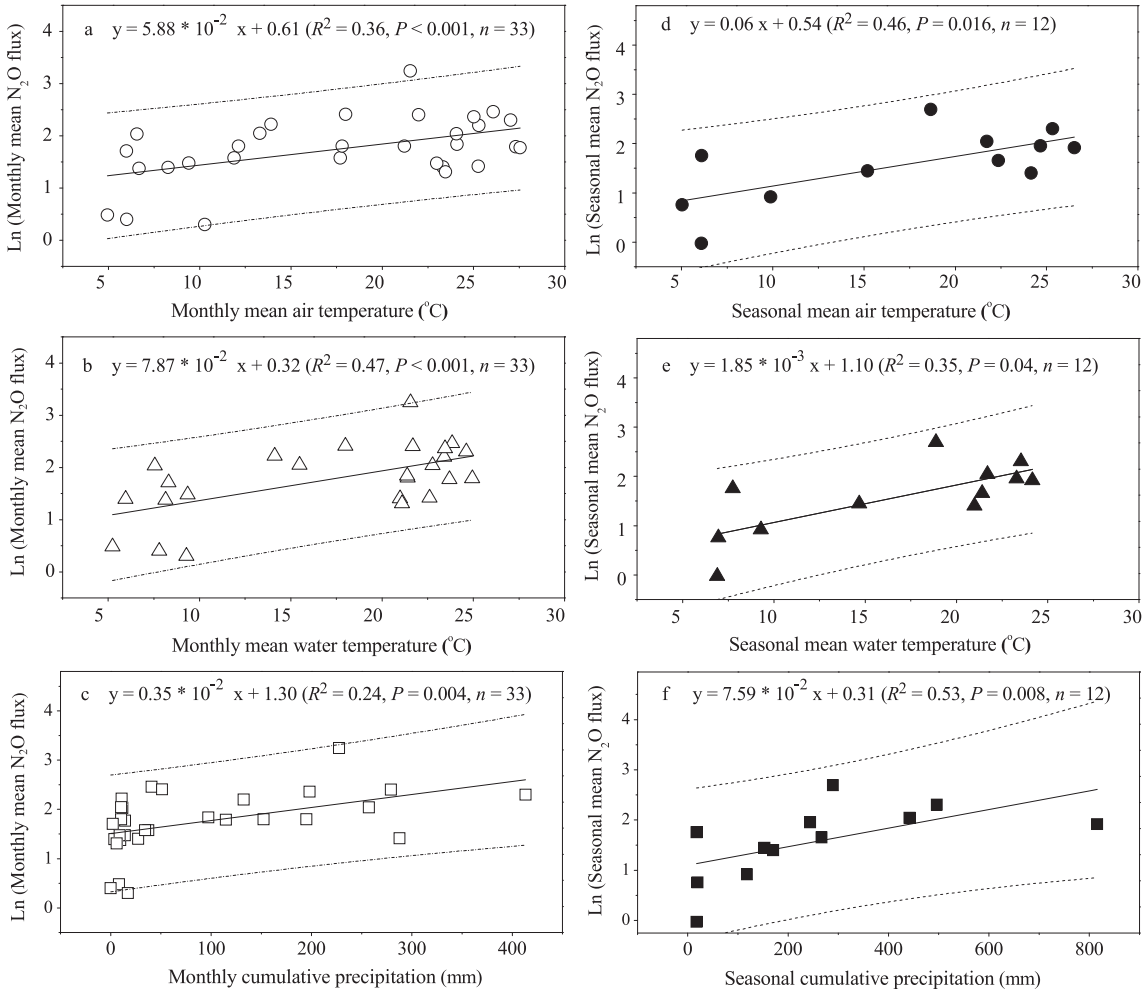

Fig. 6. Regression analyses of ln (mean monthly N_2O flux) and **(a)** mean monthly air temperature; and **(b)** mean monthly water temperature; and **(c)** monthly precipitation, and ln (mean seasonal N_2O flux) and **(d)** mean seasonal air temperature; and **(e)** mean seasonal water temperature; and **(f)** seasonal precipitation. Dashed lines are 95% confidence intervals.

reported before 2017 (Table 2), and found the mean indirect N_2O emission factor (EF_{5g} = 0.03%) in the current study was at the lower end of the range (0.01%–4.76%), although the reason for the relatively low EF_{5g} value in our study was not clear. The mean EF_{5g} (0.51%) from the literature (median: 0.18%, 95% confidence interval: 0.24%, n = 70) was higher than the default value (0.25%) proposed by the IPCC in 2006. Although the number of studies on EF_{5g} has increased since publication of the 2006 IPCC guidelines, studies on these indirect N_2O emissions remain limited (Clough et al., 2007a), and there is uncertainty about the validity of the default EF_{5g} values and the application of a single value to all drainage waters and groundwaters, due to highly variable EF_{5g} values at regional and global scales (Jurado et al., 2017, Table 2). We suggest, more *in situ* observations of indirect N_2O emission and EF_{5g} are needed to improve the reliability and confidence in the use of EF_{5g}.

4.2. Seasonal variations of N_2O emissions and affecting factors

The increase of intensive rainfall amount resulted in an increase in the level of NO_3^--N in the drainage ditch mainly receiving interflow water (Figs. 2a and 5a), confirming that rainfall affects N leaching and NO_3^--N pollution in subsurface runoff in this study area (Zhu et al., 2009; Wang and Zhu, 2011). In addition, the mean N_2O fluxes increased with cumulative monthly and seasonal precipitation (Fig. 6c and f). Our results suggest that seasonal precipitation patterns contribute to seasonal variations in NO_3^--N levels and N_2O fluxes in drainage ditches mainly receiving interflow water, which is consistent with the relationships of seasonal distributions of precipitation and the seasonal variations in NO_3^--N and N_2O emissions that have been reported from other agricultural headwater streams (Royer et al., 2004; Beaulieu et al., 2009).

In our study, the drainage ditch water was rich in NO_3^--N and DOC, and may have contained sufficient substrates for the occurrence and stimulation of denitrification (Stow et al., 2005; Beaulieu et al., 2009; Werner et al., 2012; Jahangir et al., 2013). Our results showed that the concentration of DOC in the drainage water was relatively high (Table 1) and was not related to N_2O emissions, indicating that DOC was not a limiting factor for seasonal variations in N_2O emissions. However, the concentration of NO_3^--N in the drainage water showed a positive relationship with the N_2O emission (Fig. 5c), which was similar to findings that both denitrification and N_2O production rates increase with changes in groundwater chemical characteristics, such as increasing the water NO_3^--N concentrations (Herrman et al., 2008; Beaulieu et al., 2009, 2011; Jahangir et al., 2013; Jurado et al., 2017). Denitrification has been shown to be active in generating N_2O in groundwater or areas rich in organic matter where groundwater occurred (Koba et al., 2009; Jurado et al., 2017). The studied drainage ditch was

Table 2
Comparison of EF_{5g} and indirect N_2O fluxes from drainage water, groundwater and springs reported from previous studies with dominant land use type of cropland.

Country	Source*	EF_{5g} (%)	N_2O fluxes (μg N m^{-2} h^{-1})	Reference
		Mean (range)	Mean (range)	
UK	DW	(~0.1–1.00)[c]	/	Dowdell et al. (1979)
Japan	DW	0.20 (0.059–0.44)[b]	/	Minami and Fukushi (1984)
Japan and USA	GW	0.22 (~0.01–1.00)[c]	/	Ueda et al. (1993)
UK	GW	0.25 (0.10–0.43)[b]	0.57[a]	Mühlherr and Hiscock (1997)
	SP	0.50 (0.49–0.51)[b]	/	
USA	GW	0.13 (0.06–0.19)[c]	/	Verchot et al. (1997)
UK	GW	0.34[b]	/	Mühlherr and Hiscock (1998)
	GW	0.56[b]	/	
	GW	0.04[b]	/	
Japan	GW	0.05[c]	/	Hasegawa et al. (2000)
USA	GW	0.03[b]	0.06[a]	McMahon et al. (2000)
Japan	DW	1.27[c]	/	Sawamoto et al. (2002)
UK	GW	0.19[a]	0.85[a]	Hiscock et al. (2003)
	DW	0.01 (0.005–0.028)[d]	(~100–1000)[c]	Reay et al. (2003)
Japan	DW	0.26[c] (0.076–1.05)[a]	8.53[a]	Sawamoto et al. (2003)
UK	DW	0.20[a]	/	Reay et al. (2004a)
	DW	0.06[d]	/	Reay et al. (2004b)
Japan	GW	0.15[c]	/	Sawamoto et al. (2005)
	DW	0.18[c]	/	
	DW	0.01[c]	/	
	DW	0.05[c]	/	
Germany	DW	2.41 (0.30–4.50)[b]	/	Well et al. (2005a)
	GW	0.08 (0.002–1.58)[a]	/	Well et al. (2005b)
China	GW	0.11 (0.004–0.51)[a]	/	Xiong et al. (2006)
USA	DW	0.57 (0.01–4.07)[b]	35.2 (−8.9–266.8)[a]	Beaulieu et al. (2008)
Germany	GW	4.76 (0.007–51.0)[a]	/	Weymann et al. (2008)
	GW	0.21 (0.011–1.04)[a]	/	
	GW	0.81 (0.071–7.36)[a]	/	
	GW	2.38 (0.005–24.0)[a]	/	
USA	GW	0.22 (0.13–0.31)[a]	/	Kim et al. (2009)
	GW	0.41 (0.28–0.54)[a]	/	
UK	DW	0.30 (0.008–3.60)[a]	0.87[a]	Reay et al. (2009)
Australia	GW	2.60[b]	/	Woodward et al. (2009)
Canada	DW	0.08 (0.06–0.09)[a]	9.09[d]	Baulch et al. (2011, 2012)
Italy	GW	0.15[b]	4167[d]	Laini et al. (2011)
Ireland	GW	0.73[b]	/	Jahangir et al. (2012)
UK	DW	0.61[a]	338[a]	Outram and Hiscock (2012)
France	GW	0.12[b] (0.04–0.26)[a]	0.40[a]	Vilain et al. (2012)
Germany	GW	0.35[b]	/	Well et al. (2012)
	GW	0.20[b]	/	
Ireland	GW	0.39[a]	1.71[a]	Jahangir et al. (2013)
	GW	0.41[a]	0.80[a]	
	GW	0.30[a]	1.94[a]	
	GW	0.29[a]	2.74[a]	
USA	GW	4.40 (0.2–70.0)[a]	119[b]	Gardner et al. (2016)
USA	GW	2.07[b] (0–19.9)[a]	/	Hinshaw and Dahlgren (2016)
Sweden	DW	0.17 (0.08–0.29)[d]	127.4[b] (5.5–617)[a]	Audet et al. (2017)
UK	DW	0.12 (0.003–1.06)[a]	0.57[a]	Hama-Aziz et al. (2017)
Ireland	DW	0.06[b]	/	McAleer et al. (2017)
	DW	0.01[b]	/	
	GW	0.35[b]	/	
	GW	0.05[b]	/	
New Zealand	DW	0.01[a]	/	Premaratne et al. (2017)
China	DW	0.03 (0.004–0.19)	6.56 (−0.33–40.30)	This study
Average of the EF_{5g}		0.51[AV]	/	
IPCC default value in 2006		0.25	/	IPCC (2006)

* DW: drainage water; GW: groundwater; SP: spring.
[a] Data were directly provided in the original publication.
[b] Data were recalculated from those in the original publication.
[c] Data were derived from a secondary source.
[d] Approximate value.
[AV] Average of the EF_{5g} from the reference and this study (range = 0.01%–4.76%; median = 0.18%; $n = 70$, standard error = 0.12%, CV = 1.94).

similarly rich in organic matter (7.43 g kg^{-1} soil) and DOC (mean: 41.2 mg kg^{-1} soil, range: 19.0–93.0 mg kg^{-1} soil). In addition, the DO concentration of the ditch water during the second half of the third observation year (5.58–8.57 mg L^{-1}, data not shown) was lower compared with that (7.5–15.5 mg L^{-1}) reported by Clough et al. (2007b), where coupled nitrification–denitrification could occur; moreover, the main form of inorganic N was NO_3^--N rather than NH_4^+–N in the present study, which incurred that nitrification was much less likely to occur under this condition. Therefore, we suggest that the NO_3^--N concentration was a primary factor affecting N_2O fluxes produced via denitrification, which may explain the coherence of the seasonal changes in NO_3^--N concentrations and N_2O fluxes.

Our data also revealed a positive relationship between monthly

and seasonal N_2O emissions and air and water temperature (Fig. 6a, b, 6d, and 6e). Higher temperatures induce more rapid rates of metabolism for denitrifying bacteria (Herrman et al., 2008; Jurado et al., 2017), and Stow et al. (2005) and Tian et al. (2017) found N_2O fluxes increased with increasing water temperature in downstream rivers. Moreover, increased temperature can lower the amount of dissolved N_2O in the water which in turn promotes N_2O emissions, and reduce the DO concentration leading to anaerobic conditions thus promoting denitrification (Harrison et al., 2005; Clough et al., 2007b, 2011). Besides, significant diel changes in water temperatures and day-night oxygen fluctuations could also impact N transfer such as denitrification and N_2O production (Harrison et al., 2005; Clough et al., 2007b), although diel change was not measured in this study.

Our results further confirmed that indirect emissions were evidently sensitive to fluctuations of temperature and precipitation (Griffis et al., 2017). It is likely, therefore, that heavy precipitation increased leaching and associated higher NO_3^--N concentrations in the drainage water, that when combined with warmer temperatures, resulted in stimulated denitrification and higher N_2O fluxes in summer and autumn.

Besides, we found a lack of inter-annual variation in indirect N_2O emissions, which may have been related to the lack of variation in DOC and inorganic N concentrations in the drainage ditch (Table 1), as a result of consistent annual mean temperatures, and management practices of the adjacent farmlands among the three study years. In contrast, some studies such as those in the US Corn Belt (e.g. Griffis et al., 2017) found inter-annual variations in indirect emissions attributed to warmer & wetter conditions. In the present study, 2012–2013 observation year was wetter than the other two years (precipitation: 1272 mm *vs.* 821 and 956 mm; Section 3.1), which might affect surface runoff and groundwater (Zhu et al., 2009; Wang and Zhu, 2011; Griffis et al., 2017). Wang and Zhu (2011) found that the annual mean leaching NO_3^--N concentration from local sloping farmland could be significantly lower in the wetter year (e.g. precipitation in 2006: 860 mm) than those in dryer years (e.g. precipitation in 2004: 806 mm). Although NO_3^--N concentrations of the ditch were not measured for the first observation year (2012–2013), high precipitation might lead to lower leaching NO_3^--N concentration in the first year. However, higher precipitation did not affect indirect N_2O emissions of the first year. The integrated influences of accumulated precipitation, water chemistry, and temperature could result in the N_2O emissions to be stable between years but vary with season in the drainage ditch during study period.

5. Conclusions

Large uncertainty still remains with respect to indirect N_2O emissions from agricultural systems, particularly from intensively managed farmlands such as those in China. In the present study, we investigated indirect N_2O emissions from an agricultural drainage ditch mainly receiving interflow water discharge in an intensive farming area with high N inputs located in the central Sichuan Basin. The agricultural ditch was a source of indirect N_2O emissions, with an inter-annual mean N_2O flux of 6.56 $\mu g\, N\, m^{-2}\, h^{-1}$ and a mean EF_{5g} value of 0.03%. A review of the literature revealed that the global average value of EF_{5g} was 0.51%. Although EF_{5g} in the studied ditch was lower than the global average, it is not clear that the average figure of intensive farming area in China is actually lower than the global average, because only one previous study on EF_{5g} was available in China. Further field measurements of EF_5 to reduce uncertainty in the estimate of EF_5 are needed. There were no inter-annual variations in N_2O emissions, but there were seasonal differences, where emissions were higher in summer and autumn than in winter and spring. In this study, the seasonal variations were mainly related to seasonal dynamics of the drainage water NO_3^--N concentration, temperature, and precipitation indicating the combined effects of the interactions between these factors on these indirect N_2O emissions should be considered. Our results suggest that intensive precipitation events increase water NO_3^--N concentrations and stimulate N_2O emissions from drainage ditches mainly receiving interflow water discharge. Moreover, the higher NO_3^--N concentrations of the drainage water, when combined with warmer water temperature in summer and autumn, may have stimulated denitrification and higher seasonal N_2O emissions.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.envpol.2018.07.018.

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